French Comprehensive R&D Program to Support Proliferation Resistant Innovative Fuel Cycles

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I. INTRODUCTION

Resistance to proliferation is becoming one of the key technical challenges for existing and future nuclear energy systems. Sustainability objectives, which are essential for Generation IV nuclear energy systems, require a closed fuel cycle with advanced fuel cycle processes for an adequate resistance to proliferation. Hereafter is described the French approach to such concerns in the present and the near term LWRs which will represent a large share of the nuclear fleet in France and worldwide over the 1st half of this century. For the longer term, the Generation IV International Forum offers a unique opportunity to address these issues internationally, with a great potential of innovations in fuel and fuel cycle processes.

I.1 CURRENT AND SHORT TERM SITUATION

EDF, the French national utility, is currently operating in France a nuclear fleet of 58 PWRs producing about 400 TWh annually.

For France, nuclear spent fuel is not a waste as it still contains a huge amount of energetic products. The reprocessing of spent fuel in use since 1976 at the plant of La Hague allows to recycle plutonium and some of the uranium separated from the fission products and minor actinides that constitute the ultimate waste. This practice led to the use of MOX fuel that underpins the short and medium-term policy of spent fuel management as follows :

- Around 1,100 tons of spent fuel are unloaded every year, including 100 tons of used MOX fuel. 850 tons of uranium oxide (UO_2) fuels are reprocessed annually to recover uranium and plutonium;
- The final high-level waste, made of fission products and minor actinides, is vitrified;
- The Plutonium is recycled in the 20 PWRs of the 900 MWe series that are currently licensed for MOX use (30 % MOX fuel assemblies loading), thus leading to loading 100 tons of such MOX fuel every year in France.

If extended to multiple recycle, which would require additional capacities to load MOX fuel, this strategy would tend to stabilize the Plutonium stockpile associated with the operation of the French fleet of power reactors (at about 400 tons). Scenario studies are currently performed to check whether this stockpile offers sufficient flexibility to deploy fast neutron Generation IV systems from 2035 onward.

MOX fuel is currently licensed in France for 3 cycles only as it was initially for UO_2 fuel. Following the authorization to rise the UO2 average burn up from 33 to 48 GWd/t, EDF has developed the “MOX parity project” in order to achieve burn up parity between MOX and UOx fuels. This strategy requires to increase the Plutonium content of the MOX to about 8.6 percent, so that all Plutonium extracted from the Uranium spent fuel (reduced to 850 tons/year because of the increase in burn-up) can be recycled in the 100 tons of MOX fuel loaded in the 20 MOXed 900 MWe reactors. Thus, the separated Plutonium inventory will be stabilized by 2005 at the level needed to dynamically manage the recycling process. This strategy will even make it possible to reprocess the Uranium spent fuel in addition to the 850 tons/year that are presently left in interim storage, thus leading to convert all stored PWR spent fuel into used MOX fuel by 2015, without requiring additional storage capacities.

Since the development of its generating fleet and fuel recycling strategy, France has implemented a comprehensive set of measures – agreed on an international basis - to insure adequate proliferation resistance. In particular, strict domestic and international controls are applied to avoid any diversion of nuclear materials.

Moreover, several aspects of the present fuel cycle back-up management in France offer attractive intrinsic features regarding proliferation resistance :

- Plutonium from light water reactors spent fuel is not well suited for use in weapons and does not represent the easiest route for proliferation;
- The 20 MOX-loaded PWRs are currently zero net Plutonium producers, and will become net Plutonium consumers with the increase of the authorized fuel burn-ups beyond the present level;
- One third of the recycled Plutonium is burned, and the isotopic composition of the remaining two thirds is further degraded;
- Recycling does not only further reduce the “attractiveness” of Plutonium for proliferating activities, it also enables to concentrate it in spent MOX fuels, which exhibit a higher and longer lasting radiological barrier;
- Reprocessing and recycling avoid the final underground disposal of 1100 metric tons of spent fuel, and the associated proliferation risks for future generations.

I.2 MID-TERM EVOLUTION : LWRS AND PU RECYCLING

For the mid term evolution, at least over the first half of the 21st century, LWRs will continue to play a dominant role. Indeed, Generation IV fast neutron energy systems, that afford an efficient use of natural Uranium resources and minimization of the long-lived waste production, will not be ready for a massive deployment before 2030 - 2040.

Until Generation IV energy systems are available to recycle the Plutonium generated by the current generation of LWRs, the CEA developed a specific interest in technologies enabling to reuse the Plutonium from the spent MOX fuel in existing and Generation III reactors (typically EPR).

The rationale to investigate advanced Plutonium fuels and the associated fuel cycle processes stems from the incentive to avoid accumulating dormant stockpiles of Plutonium containing spent fuels, and thus minimize both risks of proliferation and potential allocation of massive amounts of actinides to an underground repository. Furthermore, beyond the stabilization of the separated Plutonium inventory and the concentration of Plutonium in MOX spent fuels, the stabilization of the Plutonium stockpile – at a level necessary to deploy Generation IV fast neutron systems – is the next desirable objective from the viewpoints of use of natural resources, management of waste and minimization of proliferation risks.

One of the new concepts of Plutonium fuel considered is derived from current plutonium fuel technologies. It consists of islands of standard uranium dioxide (UO_2) rods surrounded by two rows of MOX fuel rods

(Figure 1). A more advanced concept consists of an heterogeneous bundle including standard UO_2 fuel rods and annular fuel elements made of plutonium oxide embedded in an inert matrix.

Figure 2 shows an example of evolution of the Plutonium isotopic vector in PWRs as a function of the burn-up. The multiple recycling reduces the fraction of fissile material, and increases the critical mass, owing to the build-up of minor actinides, both effects being unfavorable to the misuse of recycled PWR spent fuel for proliferating activities.

II. NON PROLIFERATION CRITERIA AND REQUIREMENTS FOR THE FUTURE FUEL CYCLES

Goals for Generation IV Nuclear Energy Systems have been identified and widely shared internationally :

- Improved economic competitiveness, both in terms of generating and investment costs;
- Enhanced reliability and safety in terms of operating conditions, core damage frequency, and potential consequences of off-site effects;
- Enhanced sustainability in terms of effective use natural Uranium resources and minimization of long-lived radioactive waste.

Additional objectives regarding proliferation resistance and physical protection (PR&PP) come in addition to above goals. These objectives have been defined as follows by the Generation IV International Forum :

- ***Proliferation resistance*** is those characteristics of a nuclear energy system that impede the diversion or undeclared production of nuclear material, or misuse of technology, by States in order to acquire nuclear weapons or other nuclear explosive devices;
- The ***proliferation resistance goal*** for Generation IV nuclear energy systems is to be the least desirable route to proliferation by virtue of enhanced intrinsic technical features intended to prevent or inhibit diversion or undeclared production of weapon-usable nuclear materials;
- ***Physical protection*** robustness is those characteristics of a nuclear energy system that impede the theft of materials suitable for nuclear explosives or radiation dispersal devices, and the sabotage of facilities and transportation, by sub-national entities;
- The ***physical protection goal*** for Generation IV nuclear energy systems is to be the least desirable route to theft or sabotage.

These definitions have been the starting point to assess criteria and metrics to assess how a closed fuel cycle with the associated processing technologies can be made resistant to proliferation. This work, which is in progress, led to the classification of these criteria and metrics in two families :

- Barriers : either intrinsic or extrinsic ; the objective is to incorporate within the fuel cycle design features that eliminate material acquisition paths and/or facilitate practical safeguards implementation;
- Strategic values : this consists in correlating the nature of materials at all stages of the fuel cycle with their attractiveness for proliferating activities, accounting for the isotopic composition, the radiochemical impurities and the chemical form of the fissile materials.

As the implementation of the international R&D effort for the "viability phase" of the Generation IV systems is at its very beginning, this is a unique opportunity to refine PR&PP criteria and methodology of use. This includes:

1. Adopting a comprehensive PR&PP strategy
2. Implementing measures from early design stages to operation
3. Taking benefit from the experience with safety methodology
4. Taking advantage from new technologies
5. Achieving a global optimisation of the future systems (reactor + fuel cycle)
6. Sharing the approach internationally.

The analysis led to propose the following guidelines to make the fuel cycle more resistant to proliferation risks, while ***preventing the diversion of nuclear materials*** and ***implementing safeguards*** :

- Use of radiation field : Nuclear materials should require remote handling at many stages of the process;
- Difficult misuse of facility : Any facility involved in the fuel cycle should be such that any modification would be very difficult and require specialized skills, equipment and knowledge. Environmental "signatures" associated with any facility misuse should also be looked for;
- Access to material : Nuclear material paths through the plant must be limited as much as possible and restricted access to nuclear materials must be inherent to the process, which appears to be best obtained by continuous operation. Indeed, when a process is running into continuous mode it is impossible to divert any part of the material without inducing a strong perturbation to the process itself that will be detected by the monitoring system;
- Implementation of safeguards : Use of advanced verification techniques, minimizing the interference with the routine operation of the facility;
- Implementation of safeguards : Use of built-in systems for unattended measurements made of remote monitoring devices and on-line analysis methods.

III. FUEL CYCLES STRATEGIES AND NON-PROLIFERATION

Sustainability goals call for fast neutron spectra (to transmute nuclear waste and to breed fertile fuel) and for an integral recycling of actinides from the spent fuel (plutonium and minor actinides), which fully contributes to minimize risks of proliferation. New applications, such as Hydrogen production, and economic competitiveness call for high temperature technologies (850°C), that afford high conversion efficiencies and hence less radioactive waste production and discharged heat. These orientations require breakthroughs beyond light water reactors.

In this context, the CEA decided to preserve the expertise gained on sodium cooled fast neutron reactors and to focus prospective R&D work on the development of a consistent set of gas cooled nuclear systems ranging from medium term reactor projects for electricity generation and other applications (Hydrogen production at very high temperature, actinides burning…) to a longer term vision of sustainable nuclear systems using fast neutrons with a closed and integrated fuel cycle.

The fuel cycle, which is essential to reach sustainability goals of natural resource utilization and radioactive waste minimization, is also crucial for proliferation resistance, which led to consider it, together with the reactor and the fuel, as an integral part of nuclear energy systems to be considered and optimized as a whole.

Both advanced versions of gas cooled nuclear energy systems are being substantiated by the Very High Temperature Reactor (VHTR) and the Gas fast Reactor (GFR) selected in the Generation IV International Forum.

III.1 CORE DESIGN

CEA studies on the GFR system aim at the following performances :

- Fast neutron spectrum,
- High power density (50 to 100 MW/m^3_{core}),
- High temperature of the primary coolant (480-850°C) with a pressure of 70 bar,
- Fuel operating temperature about 1250 °C in normal conditions and 1600 °C as maximum in accidental conditions,
- Self-regeneration of fissile materials,
- Full recycling of actinides.

Taking proliferation concerns into account leads to consider avoiding breeding blankets in a first step. Core optimization studies have shown that this can be afforded provided that the core contains a high volumetric fraction of heavy nuclides (> 50 %). Avoiding the need to produce plutonium in surrounding fertile blankets constitutes a strong intrinsic barrier. Means to make proliferation resistance comply with the use of fertile blankets and with a simple fuel cycle with integral recycling of transuranic elements, will be addressed in a second step to enhance the GFR breeding capability.

Furthermore, the fuel cycle processes considered processing the GFR spent fuel and re-fabrication fuel to be multi-recycled are desired to be as compact and simple as possible to partition all actinides and fission products in the minimum number of operation, with a minimum amount of actinides in the ultimate waste (so that its potential radio-toxicity be kept at a very low level). In return, the GFR system can accommodate a few percents of fission products without substantial penalty on the neutronic behavior. This concept of fuel cycle, so called "dirty fuel and clean waste" concept, is attractive in terms of proliferation resistance since it drastically enhances both the intrinsic and extrinsic barriers of the fuel cycle :

- The mixing with minors actinides readily increases the spontaneous neutron generation rate and the decay heat of the nuclear material (as illustrated on Figure 3 and Figure 4), which strongly complicates the use for a nuclear weapon;
- The very limited amount of actinides in the final waste (typically less than 0.1 %) simplifies the implementation of international safeguards on a waste repository and ensures the fact that this repository will not be, in the long term, transformed in a potential "plutonium mine".

III.2 FUEL CONCEPTS

As regards the GFR fuel element, the following requirements are envisioned :

- a typical equilibrium composition of the fuel such as : 18-20% Pu + 78-80% U + 2% M.A.,
- a high heavy atom density in the core achieved with a high fuel volume fraction (greater than 25 vol%) and a dense fuel compound such as carbide or nitride,
- a fuel capable of efficient fission product retention up to temperatures around 1600°C,
- an average power density of 100-200 MW/m^3 of fuel,

- a fast fluence in the range of 1. 10^{27} $n.m^{-2}$ (E > 0.1 MeV),
- a fission rate greater than 5% FIMA in a first step (leading to a residence time of 3 x 441 efpd) and above 10 % in the longer term.

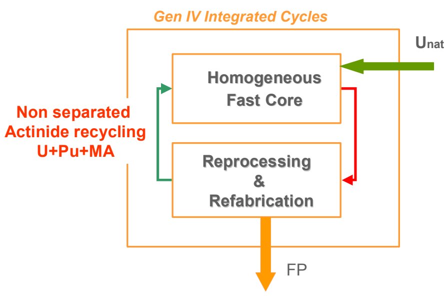
These characteristics led to consider innovative fuel concepts based on the actinides elements (kernels or fibers) coated with inert ceramics with a typical volume ratio of 70/30 % or 50/50 %. These elements are packed together into massive fuel elements though the use powder metallurgy techniques. This composite fuel aims at achieving the actinides volume fraction needed for fast neutrons, and to preserve some of the attractive features of coated particles such as the local confinement of fission products and resistance to very high temperatures. Several candidates are considered to make the inert material structure : SiC, ZrC, TiN…

In order to meet these challenging objectives, the composite structure of the fuel is adapted to reduce the effect of damage on the coating materials : the actinide elements have a typical diameter of several hundred of micrometers and the minimum distance between two fissile inclusions is much larger than twice the average fission products recoil range (i.e.: 8 – 12 µm).

Besides all improvements expected from this fuel concept in normal and abnormal service conditions, additional benefits are expected in the field of proliferation resistance, owing to the more specific head-end processing techniques needed for this composite fuel technology.

III.3 INTEGRATED FUEL PROCESSING

Besides recycling all actinides and processing these actinides in a single stream, a third feature is considered to make the GFR system more resistant to proliferation. This consists in an "integral fuel cycle", with the fuel cycle facilities integrated on the production site. On-site spent fuel processing and re-fabrication for recycling aims at minimizing transportations of nuclear materials between fuel cycle plants and the power plant. Limiting the need of transportations for the operation of the power plant to depleted Uranium as make-up fuel and packaged fission products as ultimate waste, drastically decreases the risk of nuclear material diversion and can be considered as an extrinsic barrier. This approach needs further economic assessments which certainly depend upon the national context.



Integrated Recycling & Proliferation Resistance

CEA is developing several options for the considered integrated fuel processing technologies. They can be classified into aqueous processes, pyro-metallurgical processes, and hybrid processes making use of both options, such as dissolution in molten salt and separation in aqueous solutions.

III.3.1 Hydrometallurgical processes :

Relying on a large industrial experience, these techniques show excellent performances and induce a low amount of secondary waste. Among the drawbacks that call for improvements for the GFR fuel cycle, are the poor compactness of today's equipments and the sensitivity to radiolysis of organic solvents used to perform the An – FP separation step.

One of the major challenges is to develop a grouped actinides separation pathway, which is already under way. This task is made difficult by the fact that all current actinides separation processes are based on the redox properties of these elements in acidic aqueous media. Indeed the stable oxidation state is 6 for Uranium, 5 for Neptunium, 4 for Plutonium and 3 for Americium and Curium. Thus any of the classical chemistry processes cannot separate these actinides altogether from the fission products, and specific complexants and/or redox reagents must be identified and developed. Figure 5 describes a first flow-sheet that presents real features of proliferation resistance and is currently under investigation.

III.3.2 Pyrometallurgical processes :

These processes, although they never gained industrial maturity, have several known (or presumed) advantages:

- They are well-suited for metallic fuels as it has been demonstrated by ANL with the EM process;
- Their ability to treat refractory materials as those described above should be high;
- They can handle high burn-up fuels thanks to the radiochemical stability of molten salts;
- They allow larger loadings of fissile materials, as the constraints of criticality are less restrictive than those of aqueous processes which contain hydrogenous materials.

However, today's know-how is not sufficient and some drawbacks & uncertainties must be worked on :

- Separation performances;
- Corrosion;
- Waste form and amount;
- Sophisticated technology (High temperature, controlled atmosphere).

The is currently conducting an R&D program to assess pyrochemical processes and related technologies along two main routes for a grouped actinides recovery: the concept of liquid cathode electrolysis based on the flow-sheet initially proposed by Argonne National Laboratory (Figure 6) and the liquid-liquid extraction between a molten salt and a fused metal. In that domain, very promising result have already been obtained in terms of grouped separation. Figure 7 shows the conceptual flow-sheet of a process currently under development at CEA based on the extraction of the actinides by an aluminum-cupper alloy from a fluoride melt.

IV. CONCLUSIONS

In collaboration with other member countries of the Generation IV International Forum, France has launched a comprehensive research program on advanced fuel cycle processes aiming at making closed nuclear fuel cycles compatible with an adequate resistance to proliferation. Closed fuel cycles are needed for sustainable nuclear energy systems to preserve natural Uranium resources (while recycling fissile isotopes) and to minimize the environmental impact (while recycling the long-lived minor actinides for transmutation). Their cost should be kept acceptable (looking for straight forward processes with compact technologies) and as resistant as possible to proliferation at any step. This is being achieved, while considering the following guidelines, that are especially considered for the Generation IV systems, and the GFR in particular:

- *When thinking of a separation:* Look for processes that do separate actinides as a group
- *When developing a separation process*: Favor continuous operation
- *When engineering a separation process*: Ease the implementation of safeguards.

The program is currently being implemented, with a major emphasis put on fuel particles for the VHTR and on the innovative fuels envisaged for the GFR. As for other Generation IV systems, fuel and the fuel cycle are integral parts of the nuclear system to be optimized consistently, while providing adequate resistance to proliferation. The best use is made of advanced technologies in the field of composite ceramics outside the nuclear sector as well as basic research to resolve the feasibility issues of the innovative fuels considered. This program will also require irradiation testing in representative conditions and will take best benefit from the experimental capabilities of the hot laboratory Atalante to allow for the construction of a GFR demonstration facility around 2020/2025.

Until Generation IV fast neutron systems are deployed, Plutonium recycle is made on an industrial basis in French operating PWRs and further steps are taken to enable a few steps of recycle to avoid accumulating dormant stockpiles of Plutonium containing spent fuels, and thus minimize both risks of proliferation and potential allocation of massive amounts of actinides to a geological repository.

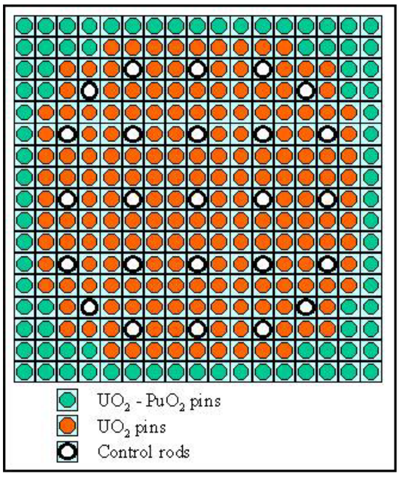


Figure 1. - Schematic drawing of a CORAIL assembly proposed for multiple recycling of Plutonium in PWRs.

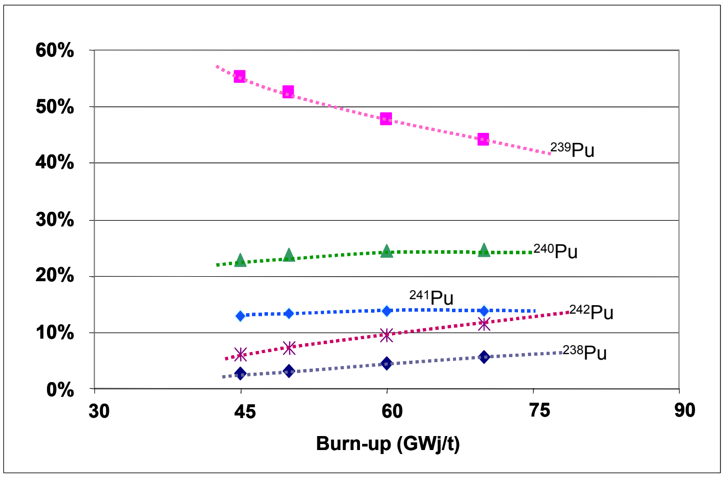


Figure 2. - Influence of the burn-up on the plutonium isotopic composition for a PWR UO_2 fuel (Initial Enrichment: 4.5%, cooling time: 3 years).

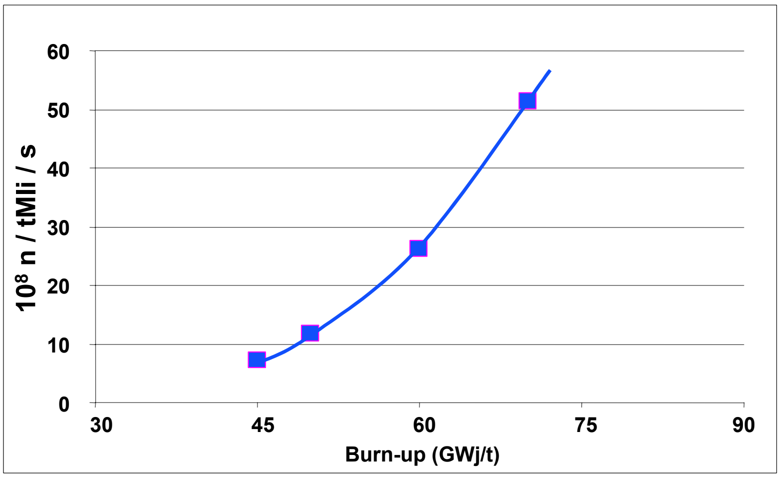


Figure 3. - Influence of the burn-up on the neutron emission rate of actinides for a PWR UO_2 fuel (Initial Enrichment: 4.5%, cooling time: 3 years).

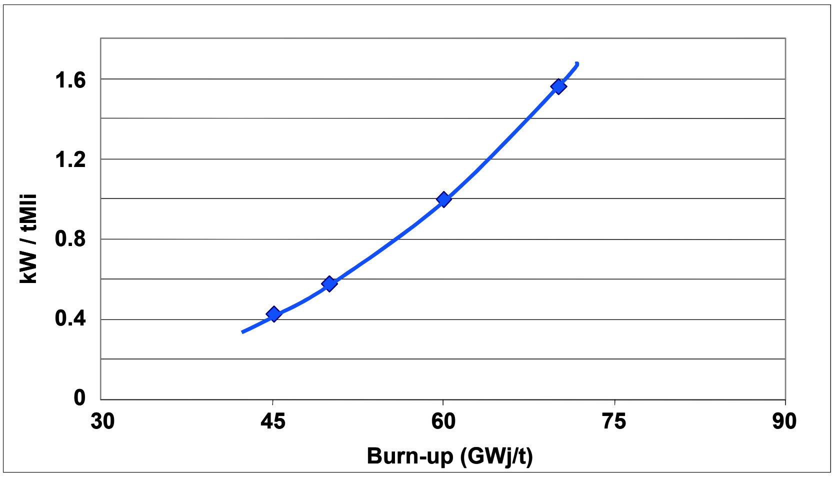


Figure 4. - Influence of the burn-up on the Actinides specific power for a PWR UO_2 fuel (Initial Enrichment: 4.5%, cooling time: 3 years).

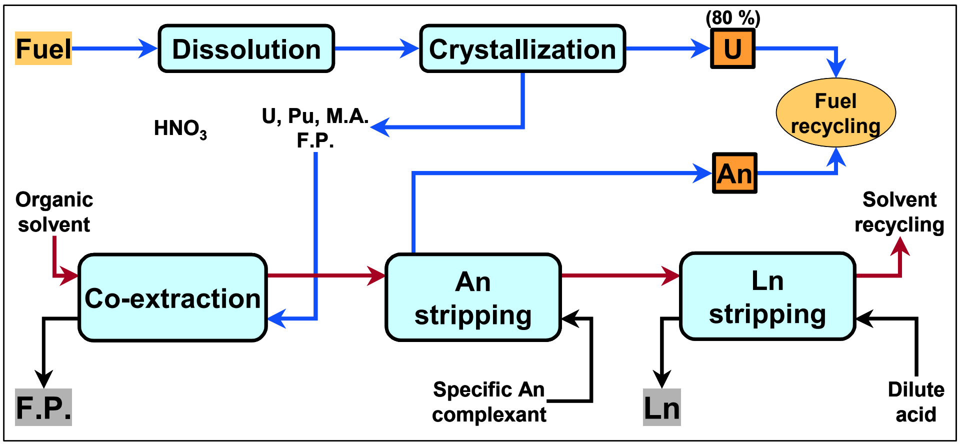


Figure 5. - The GANEX concept : Group ActiNides EXtraction.

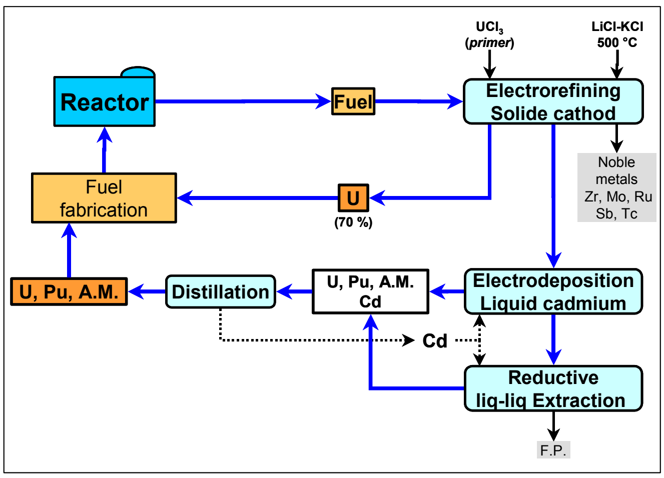


Figure 6. - Recovery of the actinides with the electrometallurgical (EM) process based on the ANL concept for the IFR fuel cycle.

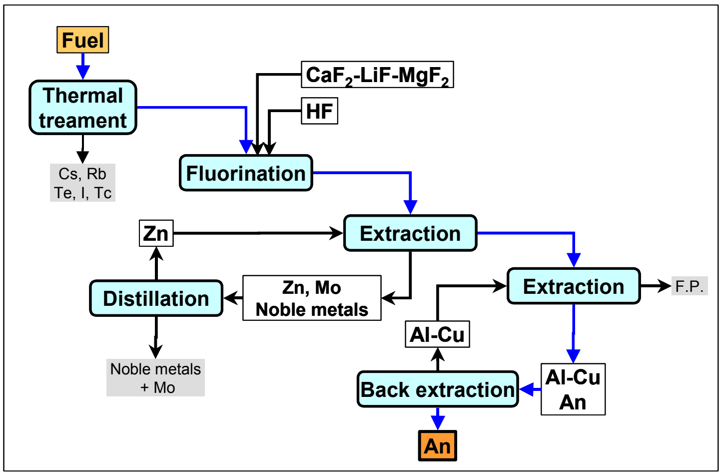


Figure 7 . Group actinides separation with the PYREX process (PYrochemical treatment by Reductive EXtraction).